

Poly(sulfobetaine)s and Corresponding Cationic Polymers. VII. Thermal Degradation of Copolymers Derived from Poly(Acrylamide *co-N,N*-Dimethylaminopropylmaleimide)

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ABSTRACT: The copolymer prepared by copolymerizing acrylamide and maleic anhydride was imidized with *N,N*-dimethylaminopropylamine. The obtained acrylamide-*N,N*-dimethylaminopropylmaleimide (ADMAMP) copolymer was then reacted with propane sulfone to yield an acrylamide-*N,N*-dimethylmaleimidopropyl ammoniopropyl sulfonate (ADMMAPS) copolymer or then reacted with methyl iodide to yield a poly(methyl iodide quaternized acrylamide-*N,N*-dimethylaminopropyl maleimide) copolymer [poly(MIQADMAMP)]. The kinetic parameters, such as the reaction order, the activation energy, and the preexponential factor, of the thermal degradation were evaluated from thermal gravimetric curves for these two copolymers. The activation energy and preexponential factor of the poly(ADMAMP) were higher than those of poly(MIQADMAMP) for Ozawa's method and van Krevelen's method. © 1997 John Wiley & Sons, Inc. *J Appl Polym Sci* **66**: 95–103, 1997

Key words: acrylamide-*N,N*-dimethylmaleimidopropyl ammoniopropyl sulfonate copolymer; poly(methyl iodide quaternized acrylamide-*N,N*-dimethylaminopropylmaleimide); thermal degradation

INTRODUCTION

Sulfobetaine monomers derived from dimethylaminoalkylacrylates and dimethylaminoalkylacrylamides are widely used in industries concerned with textiles, dispersion agents, medical products, antistatic agents, surfactants, protective colloids, and other related materials.^{1–12} The aqueous solution properties of poly(betaines), such as carboxybetaine and sulfobetaine, have been extensively investigated.^{13–27}

A series of poly(sulfobetaine)s and corresponding cationic polymers prepared from zwitterionic sulfobetaine and corresponding cationic mono-

mers were studied previously in our laboratory.^{28–37} The aqueous solution properties of cationic poly-(trimethylacrylamidopropylammonium iodide) [poly(TMAAI)], poly(*N,N'*-dimethylacrylamidopropyl ammonio propanesulfonate) [poly(DMAAPS)], and poly(*N,N'*-dimethylmethacryloxyethyl ammonio propanesulfonate) [poly(DMAPS)] were reported in previous articles.^{15,32,33} Recently, studies of polyampholytes prepared from copolymers having high purities and similar structural characteristics have been the focus of our laboratory. For example, a styrene-*N,N*-dimethylmaleimidopropyl ammonio propanesulfonate (SDMMAPS) copolymer was prepared from a styrene–maleic anhydride copolymer.³⁴ The properties of this ampholytic SDMMAPS copolymer in aqueous salt solution showed a marked distinction from sulfobetaine homopolymers, es-

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pecially poly(DMAAPS). This phenomenon is attributed mainly to the presence of the styrene segment, a hydrophobic group, in the pendent group of the SDMMAPS copolymer. Recently, the synthesis and aqueous solution properties of poly(methyl iodide quaternized acrylamide-*N,N*-dimethylaminopropyl maleimide) copolymer [poly(MIQADMAMP)] and acrylamide-*N,N*-dimethylmaleimidopropyl ammonio propane sulfonate [poly(ADMMAPS)] copolymers were studied in previous reports.^{36,37}

The thermal degradation of the poly(DMAPS) and poly(DMAAPS) has been reported in previous articles.^{28,31} The investigation of the thermal behaviors of these two copolymers, poly(MIQADMAMP) and poly(ADMMAPS), is the aim of this article. The kinetic parameters, such as activation energy, reaction order, and preexponential factor, of the thermal degradation for these two copolymers determined by Ozawa's method and van Krevelen's methods will be compared in this article.

EXPERIMENTAL

Synthesis of Alternating Acrylamide-Maleic Anhydride Copolymer

The alternating copolymer acrylamide-maleic anhydride (AA-MA; molar ratio, 1 : 1) was synthesized as previously described.^{35,36,38} The viscosity average molecular weight (\bar{M}_v), measured in 0.6*N* NaNO₃ at 25°C with a Ubbelohde viscometer, is 3.06×10^4 .³⁹

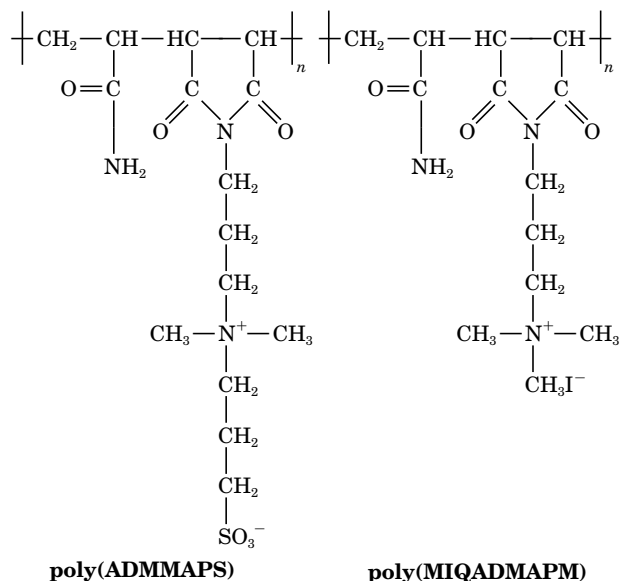
Imidization of AA-MA Copolymer

The AA-MA copolymer was imidized with 3-dimethylaminopropylamine as described in previous articles.^{35,36} The imidization degree measured by elemental analysis is 98.1 mol %.

Preparation of Poly(MIQADMAMP) and Poly(ADMMAPS) Copolymers

The copolymers [poly(MIQADMAMP)] and [poly(ADMMAPS)] were prepared as described previously.^{35,36} The copolymers were characterized by elemental analysis techniques and infrared spectra, as described before, and were quite hygro-

scopic. The poly(MIQADMAMP) was a yellow-white solid, and its intrinsic viscosity was 0.117 dL/g in a 0.1*M* NaCl solution. The degree of formation of the quaternary, calculated by elemental analysis, is 96.72 mol %. The poly(ADMMAPS) was a brown-white solid, and its intrinsic viscosity was 0.102 dL/g in a 0.1*M* NaCl solution. The degree of the formation of the sulfobetaine, calculated by elemental analysis of sulfur, is 98.05 mol %. The structures of these two copolymers are



Thermal Analysis

Thermal analysis was measured by a DuPont 2000 thermal analyzer. The heating rate was at 5, 10, 20, and 40°C/min in nitrogen atmosphere. The flow rate was 30 mL/min, and a sample weight of 10 mg was used.

RESULTS AND DISCUSSION

Thermogravimetric Curve Characteristics of Poly(MIQADMAMP) and Poly(ADMMAPS) Copolymers

The poly(MIQADMAMP) was quite hygroscopic at ambient temperature, so the thermogravimetric (TG) analysis was performed after the moisture was removed at 120°C for 5 min. The typical TG and derivative TG curves for the thermal degradation of a 10-mg poly(MIQADMAMP) sample

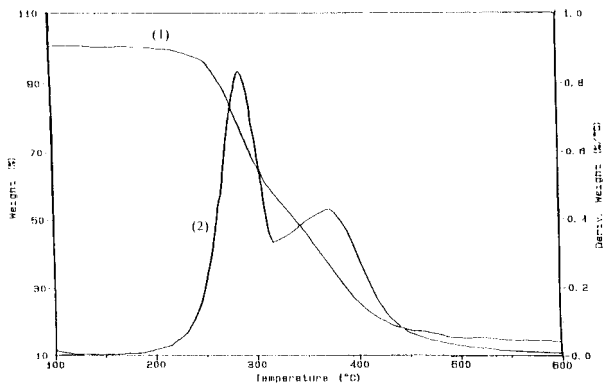


Figure 1 The curves of (1) TG analysis and (2) derivative TG analysis obtained under a nitrogen atmosphere degradation of poly(MIQADMAPM).

are shown in Figure 1, with a heating rate of 10°C/min under a nitrogen atmosphere. From Figure 1, it can be seen that the thermal degradation of such a copolymer occurs in two stages under a nitrogen atmosphere. The poly(ADMMAPS) copolymer was also treated by the same procedure as the poly(MIQADMAPM) copolymer for the TG analysis. The TG curve for the thermal degradation of a 10-mg poly(ADMMAPS) copolymer sample is shown in Figure 2, with a heating rate of 10°C/min under a nitrogen atmosphere. Figure 2 also shows that the thermal degradation of poly(ADMMAPS) occurs in two stages under a nitrogen atmosphere. This thermal behavior was also observed in our previous reports.^{28,29,31}

The TG curve for poly(MIQADMAPM) shows a two-stage thermal degradation in nitrogen atmosphere. In the first stage, weight loss starts at 247°C and continues to 302°C with 34.3% weight loss. The second stage (39.3%) is completed by 411°C. The weight percentage remaining at 600°C is 14.6%. The TG curve for poly(ADMMAPS) in nitrogen atmosphere also exhibits two stages. Weight loss starts at 264°C and continues to 314°C with 30.7% weight loss in the first stage. The second stage (34.3%) is completed by 425°C. The weight percentage remaining at 600°C is 24.5%. Tables I and II show the percent weight loss of poly(MIQADMAPM) and poly(ADMMAPS) copolymers in nitrogen atmosphere and the corresponding temperatures at heating rates of 5, 10, 20, and 40°C/min, respectively. The results shown in Tables I and II indicate that the initial decomposition temperature of the poly(ADMMAPS) copolymer is higher than

that of the poly(MIQADMAPM) copolymer and that the poly(ADMMAPS) copolymer also has a higher weight percentage of residue at 600°C at various heating rates.

Determination of Kinetic Parameters for the Thermal Degradation of Poly(MIQADMAPM) and Poly(ADMMAPS) Copolymers

Overall Apparent Kinetic Parameters Determined by Ozawa's Method

The results of the thermogravimetry at various heating rates under a nitrogen atmosphere are plotted against the temperature in Figures 3 and 4 for poly(ADMMAPS) and poly(MIQADMAPM) copolymers, respectively. The apparent activation energy was determined by Ozawa's method⁴⁰ for a given value of weight fraction (wt %). According to it, the weight decreases to a given fraction at temperature T_1 for a heating rate of β_1 , at T_2 for β_2 , and so on. The following equation could be derived:

$$-\log \beta_1 - \frac{E_1}{2.303RT_1} = -\log \beta_2 - \frac{E_2}{2.303RT_2} \quad (1)$$

where E is the activation energy and R is the ideal gas constant. From Figures 3 and 4, the logarithms of the heating rates could be plotted against the reciprocal absolute temperature until the weight of the sample decreases to a given fraction. The results are shown in Figures 5 and 6. Straight lines are drawn using the linear regression method. From these lines, the activation energies and the preexponential factors are determined, as listed in Table III for poly(MIQAD-

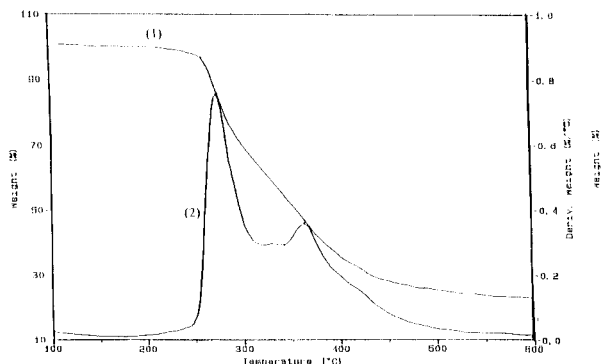


Figure 2 The curves of (1) TG analysis and (2) derivative TG analysis obtained under a nitrogen atmosphere degradation of poly(ADMMAPS) copolymer.

Table I Weight Loss of Poly(MIQADMAPM) Under a Nitrogen Atmosphere at Various Heating Rates

Heating Rate (°C/min)	First Stage		Second Stage		Weight Remaining (%) at 600°C
	Temperature (°C)	Weight Loss (%)	Temperature (°C)	Weight Loss (%)	
5	245 ~ 295	32.8	295 ~ 395	37.2	18.2
10	247 ~ 302	34.3	302 ~ 411	39.3	14.6
20	254 ~ 319	35.0	319 ~ 441	41.4	15.7
40	276 ~ 338	34.0	338 ~ 471	42.1	14.3

MAPM) and poly(ADMMAPS) copolymers. The results shown in Table III indicate that the activation energies and the preexponential factors of the poly(ADMMAPS) copolymer in the first degradation stage are approximately equal to those of the poly(MIQADMAPM) copolymer, but the activation energies and the preexponential factors of the poly(ADMMAPS) copolymer are higher than those of the poly(MIQADMAPM) copolymer in the second degradation stage. In addition, the identical kinetic parameters (E and A values) for the two successive degradation stages of the poly(MIQADMAPM) copolymer are incidentally observed in the table. Another result shows that the apparent overall activation energy and preexponential factor for the poly(ADMMAPS) copolymer are larger than those for the poly(MIQADMAPM) copolymer. This occurrence demonstrates that the degradation mechanisms of these two corresponding copolymers are different at the same conditions of degradation.

The kinetic order of the reaction, n , could be derived by using eq. (1):

$$\log |\ln(1 - C)| = \log \left(\frac{AE}{\beta R} \right) - 2.35 - \frac{E}{2.303RT} \quad \text{for } n = 1 \quad (2)$$

Table II Weight Loss of Poly(ADMMAPS) Copolymer Under a Nitrogen Atmosphere at Various Heating Rates

Heating Rate (°C/min)	First Stage		Second Stage		Weight Remaining (%) at 600°C
	Temperature (°C)	Weight Loss (%)	Temperature (°C)	Weight Loss (%)	
5	257 ~ 304	33.6	304 ~ 404	30.0	24.3
10	264 ~ 314	30.7	314 ~ 425	34.3	24.5
20	271 ~ 350	38.9	350 ~ 440	38.9	22.9
40	281 ~ 361	40.0	361 ~ 466	31.4	21.4

where A is the preexponential factor, β is the heating rate, R is the ideal gas constant, and E is the activation energy.

The conversion (C) is defined by $C = 1 - (W/W_0)$ where W_0 and W represent the initial weight and weight at any time, respectively, and n is the reaction order. The logarithm of $|\ln(1 - C)|$ can be plotted versus the logarithm of heating rate (β) for a given temperature, and the respective results are shown in Figures 7 and 8. Good linear relationships are obtained at constant temperatures of 290 and 370°C and 280 and 370°C, which are representative temperatures of the two degradation stages at the maximum degradation rate, under a nitrogen atmosphere for poly(MIQADMAPM) and poly(ADMMAPS) copolymers, respectively. These results indicate that the apparent kinetic order is unity under a nitrogen atmosphere in both samples.

Maximum Reaction Parameters Determined by Van Krevelen's Method

The activation energy was determined by the method of van Krevelen's et al.⁴¹ They reported that at a certain temperature, T_m , a maximum

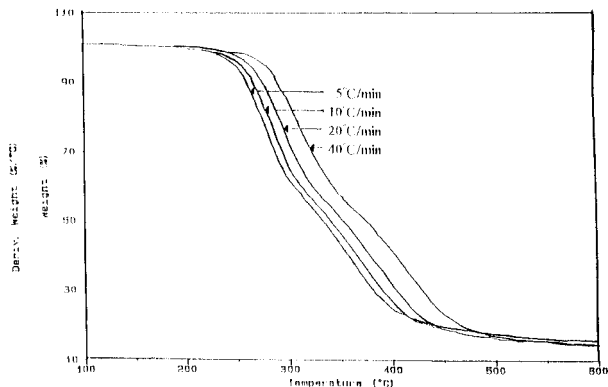


Figure 3 TG curves of poly(MIQADMAPM) at various heating rates under a nitrogen atmosphere.

reaction velocity is measured. When the reaction proceeds almost entirely within the range $0.9 T_m < T < 1.1 T_m$, the following equation could be derived:

$$\ln \int_0^C \frac{dC}{(1-C)^n} = \ln \left[\frac{A}{\beta} \left(\frac{0.368}{T_m} \right)^{E/RT_m} \times \frac{1}{\frac{E}{RT_m} + 1} \right] + \left(\frac{E}{RT_m} + 1 \right) \ln T \quad (3)$$

where E is the activation energy, R is the ideal gas constant, β is the heating rate, and A is the preexponential factor.

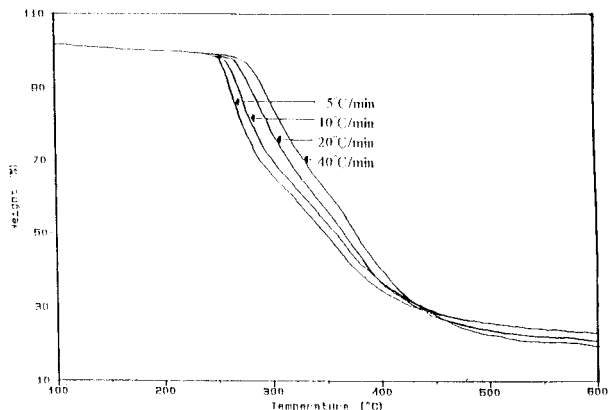


Figure 4 TG curves of poly(ADMMA)PS copolymer at various heating rates under a nitrogen atmosphere.

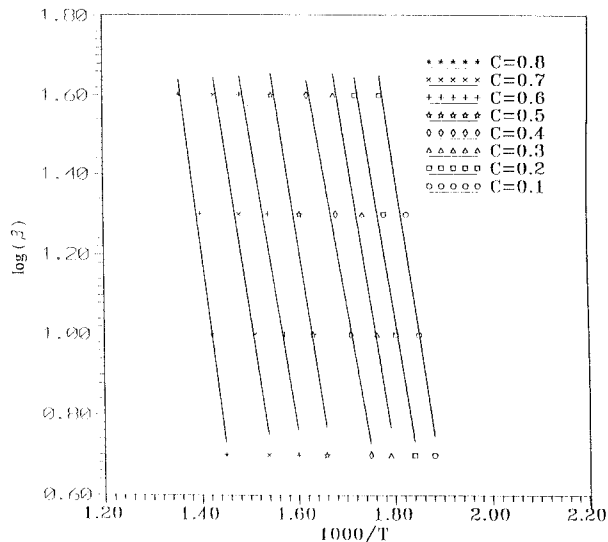


Figure 5 The plots of logarithms of heating rate versus the reciprocal absolute temperature for indicated conversions of the degradation of poly(MIQADMAPM) under a nitrogen atmosphere.

From eq. (3), the term $\left(\int_0^C \frac{dC}{(1-C)^n} \right)$ is replaced by $\ln|\ln(1-C)|$ when the reaction order (n) is unity. Hence, eq. (3) can be given by

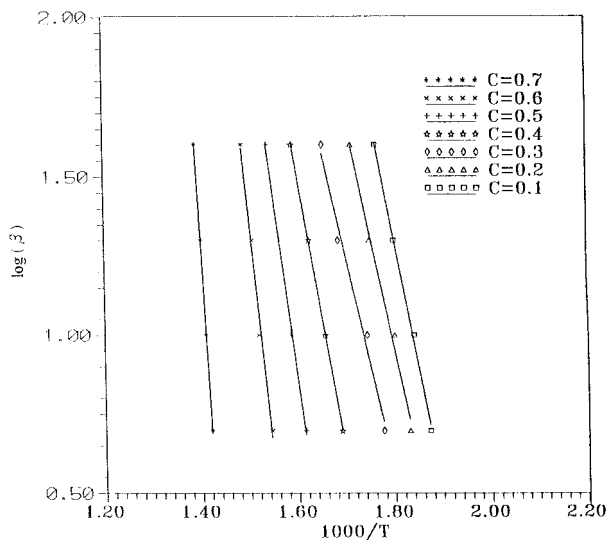


Figure 6 The plots of logarithms of heating rate versus the reciprocal absolute temperature for indicated conversions of the degradation of poly(ADMMA)PS copolymer under a nitrogen atmosphere.

Table III Activation Energies and Preexponential Factors of the Degradation of Poly(MIQADMAPM) and Poly(ADMMAAPS) Copolymers Under a Nitrogen Atmosphere by Ozawa's Method

Stage	Poly(MIQADMAPM)		Poly(ADMMAAPS)	
	E (kJ/mol)	log A ^a	E (kJ/mol)	log A ^a
First	147	15.5	146	15.1
Second	147	14.6	270	23.1
Overall average	147	15.0	208	18.6

^a The dimension of A is min⁻¹.

$$\ln|\ln(1-C)| = \ln \left[\frac{A}{\beta} \left(\frac{0.368}{T_m} \right)^{E/RT_m} \times \frac{1}{\frac{E}{RT_m} + 1} \right] + \left(\frac{E}{RT_m} + 1 \right) \ln T \quad (4)$$

From eq. (4), the natural logarithm of $|\ln(1-C)|$ can be plotted against the natural logarithm of absolute degradation temperature (T) under a nitrogen atmosphere. Figures 9 and 10, respectively, show the relationship given by eq. (4) of van Krevelen et al. for poly(MIQADMAPM) and poly(ADMMAAPS) copolymers in the first-stage degradation. The observed good linearities show that the reaction order is unity, and the acti-

vation energies and preexponential factors were calculated from the slope and intercepts, respectively, with different heating rates in a nitrogen atmosphere. Similarly, Figures 11 and 12, respectively, show the linear relations for the two copolymers in the second stage. The kinetic parameters (E and log A) in the first and second degradation stages for these two copolymers are listed in Tables IV and V. The results shown in Tables IV and V indicate that the activation energies for the poly(MIQADMAPM) copolymer remain nearly constant in both stages, but the preexponential factors are increased with increasing heating rate. For the poly(ADMMAAPS) copolymer, the activation energies in the first stage decrease with increasing heating rate and increase with an in-

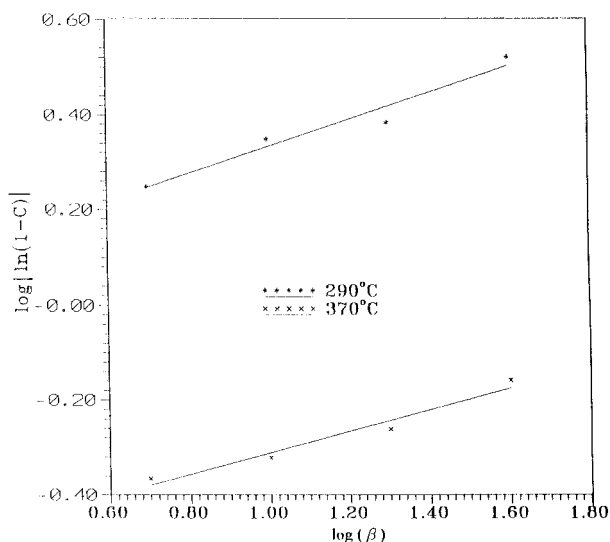


Figure 7 Relationship of poly(MIQADMAPM) obtained by Ozawa's method for $n = 1$ under a nitrogen atmosphere.

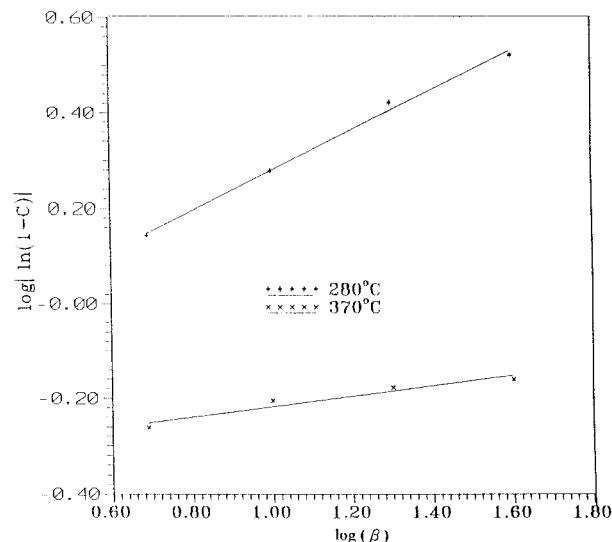


Figure 8 Relationship of poly(ADMMAAPS) copolymer obtained by Ozawa's method for $n = 1$ under a nitrogen atmosphere.

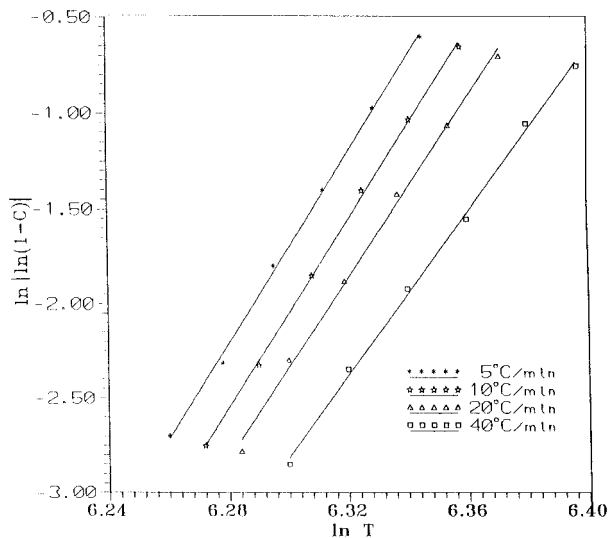


Figure 9 Application of van Krevelen's method to experimental data of poly(MIQADMAMPM) for the first thermal degradation stage under a nitrogen atmosphere.

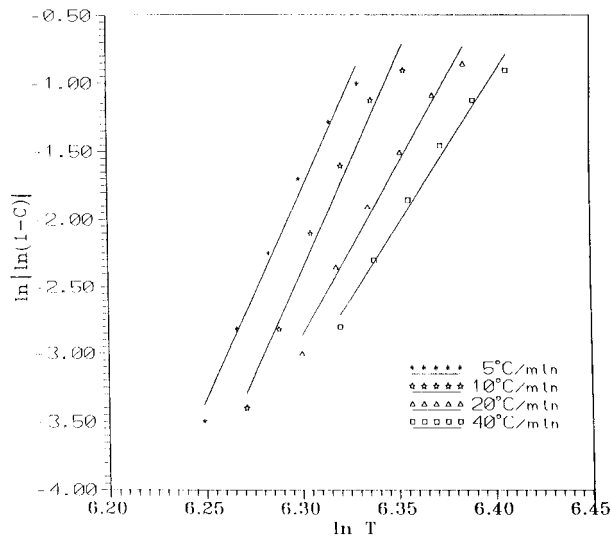


Figure 11 Application of van Krevelen's method to experimental data of poly(MIQADMAMPM) for the second thermal degradation stage under a nitrogen atmosphere.

crease of heating rate in the second stage; however, the preexponential factors increase with an increase of heating rate in both stages. Comparing Tables IV and V, the activation energies in the first degradation stage for the poly(ADMMAPS)

copolymer are larger than those for the poly(MIQADMAMPM) copolymer. The results shown in Tables IV and V also explicitly indicate that the activation energies of the maximum degradation for the poly(MIQADMAMPM) copolymer are not affected by the heating rates. However, the thermal

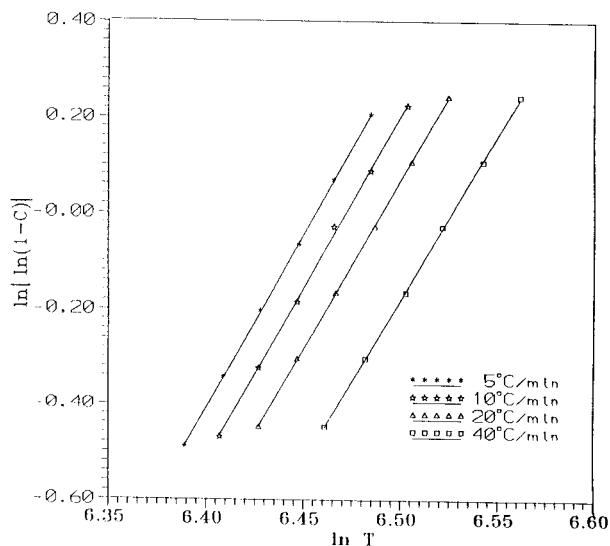


Figure 10 Application of van Krevelen's method to experimental data of poly(ADMMAPS) copolymer for the first thermal degradation stage under a nitrogen atmosphere.

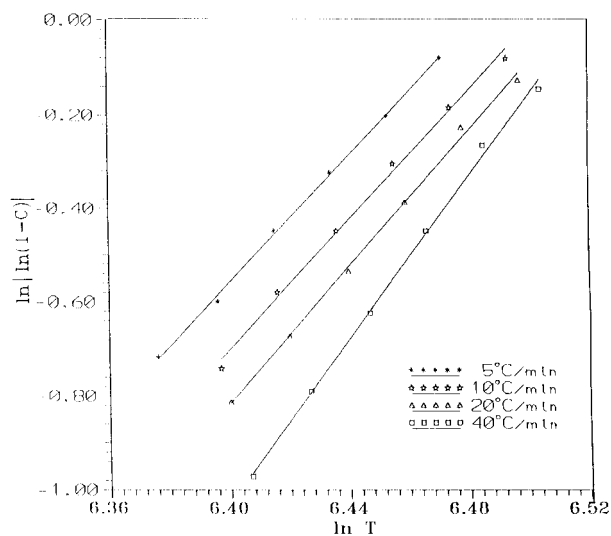


Figure 12 Application of van Krevelen's method to experimental data of poly(ADMMAPS) copolymer for the second thermal degradation stage under a nitrogen atmosphere.

Table IV Activation Energies and Preexponential Factors of First-Stage Thermal Degradation of Poly(MIQADMAPM) and Poly(ADMMAPS) Copolymer Under a Nitrogen Atmosphere by Van Krevelen's Method

Heating Rate (°C/min)	Poly(MIQADMAPM)			Poly(ADMMAPS)		
	T_m (°C)	E (kJ/mol)	log A	T_m (°C)	E (kJ/mol)	log A
5	277.9	94	10.6	262.3	168	12.3
10	285.1	97	12.3	274.3	167	13.8
20	292.2	93	13.8	291.1	121	14.5
40	306.7	96	15.4	303.1	110	15.8

degradation of the poly(ADMMAPS) copolymer is profoundly affected by the heating rates. This result was also observed in our previous article.^{29,31} Furthermore, comparing the data on E and A values obtained from the methods of Ozawa and van Krevelen, and shown in Tables III–V for a given copolymer [poly(MIQADMAPM) or poly(ADMMAPS) copolymer], the result shows a significant difference. This is due mainly to these two methods having some different assumptions and approximations.^{40,41}

On the other hand, it is interesting for us to compare the thermal degradation behavior of the two copolymers with that of their parent AA-MA copolymer. Vilcu et al.³⁹ reported that the thermal degradation of the AA-MA copolymer showed four degradation stages in TG analysis. The characteristic temperature, activation energy, and weight loss of the AA-MA copolymer (molar ratio, 1 : 1) in the respective stages are 50–120°C, 44 kJ/mol, 7.5% for stage I; 120–230°C, 68 kJ/mol, 16.5% for stage II; 230–330°C, not measured, 8% for stage III; and 330–500°C, 160 kJ/mol, 30% for stage IV. They considered that the degradation processes are mainly water loss, imidization with NH_3 elimination (intermolecular crosslinking), decarboxylation of MA, and over 420°C random scission of

the main chain. However, the data shown in Tables IV and V exhibit degradation behaviors for the two copolymers that are different from their parent copolymer's results. Hence, these results explicitly demonstrate that the thermal degradation behaviors of the two copolymers (which are different from the AA-MA copolymer) are profoundly affected by the side chains derived from the MA unit in the copolymer chain.

CONCLUSIONS

The thermal behaviors of poly(MIQADMAPM) and poly(ADMMAPS) copolymers exhibit a two-stage thermal degradation under a nitrogen atmosphere. The initial decomposition temperature for poly(ADMMAPS) is higher than that for poly(MIQADMAPM). The kinetic parameters of the thermal degradation for these two corresponding copolymers are calculated by Ozawa's method and van Krevelen's method. The overall apparent activation energy and preexponential factor for poly(ADMMAPS) are larger than those for poly(MIQADMAPM). The activation energy of the maximum degradation for poly(MIQADMAPM) is not affected by the heating rate, but that for poly(AD-

Table V Activation Energies and Preexponential Factors of Second-Stage Thermal Degradation of Poly(MIQADMAPM) and Poly(ADMMAPS) Copolymers Under a Nitrogen Atmosphere by Van Krevelen's Method

Heating Rate (°C/min)	Poly(MIQADMAPM)			Poly(ADMMAPS)		
	T_m (°C)	E (kJ/mol)	log A	T_m (°C)	E (kJ/mol)	log A
5	358.4	33	6.9	350.0	27	6.0
10	370.4	33	8.5	363.2	29	7.8
20	383.5	33	10.0	365.6	32	9.9
40	407.7	33	11.5	370.4	38	12.2

MMAPS) is profoundly affected by the heating rate.

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